Pseudogap in Doped Mott Insulators is the Near-neighbour Analogue of the Mott Gap

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We show that the strong coupling physics inherent to the insulating Mott state in 2D leads to a jump in the chemical potential upon doping and the emergence of a pseudogap in the single particle spectrum below a characteristic temperature. The pseudogap arises because any singly-occupied site not immediately neighbouring a hole experiences a maximum energy barrier for transport equal to t^2/U , where t is the nearest-neighbour hopping integral and U the on-site repulsion. The resultant pseudogap cannot vanish before each lattice site, on average, has at least one hole as a near neighbour. The ubiquituity of this effect in all doped Mott insulators suggests that the pseudogap in the cuprates has a simple origin.

In a Mott insulator with one orbital per site, each unit cell contains an odd number of particles but the Fermi energy lies in the middle of a gap. In contrast, band insulators contain an even number of electrons per unit cell and the Fermi energy lies atop a full band. Consequently, Mott and band insulators are not adiabatically connected. Nonetheless, most Mott insulators tend to order antiferromagnetically below some temperature, T_N . As a consequence, it is standard to view the Mott state simply as an antiferromagnet in which the unit cell has doubled. On this view, the insulating properties of a Mott insulator are equivalent to those of a band insulator. Band insulators possess rigid bands and hence doping only creates quasiparticles at the Fermi level. That this picture fails fundamentally in the parent cuprates, which are all antiferromagnetic Mott insulators, is immediately evident from optical conductivity experiments[1, 2] which reveal that even for $T \gg T_N$, a gap of order 2eV exists and doping leads to a massive reshuffling of spectral weight from 2eV to the Fermi energy. These experiments lay plain that what is missing in the antiferromagnetic reduction is Mottness itself: 1) in the absence of magnetic ordering $(T > T_N)$, a charge gap exists in the single particle spectrum, 2) each electronic state in the first Brillouin zone has spectral weight both above and below the charge gap, and 3) the sum rule that each singleparticle state carries unit weight is satisfied[3] only when the spectral function is integrated across the charge gap not simply up to the chemical potential as in a band insulator. A consequence of (3) is that in the Mott state, the traditional notion that the chemical potential demarcates the boundary between zero and unit occupancy fails fundamentally. This failure is central to Mottness.

The breakdown of the traditional band insulator sum rule in the cuprates is well described[3] by the Hubbard model in which the on-site energy for double occupancy leads to a charge gap at half-filling. Such local on-site physics dominates the insulating behavour at half filling. In the lightly doped regime, $\delta \approx 0$, effective interactions of longer range come into play as neighbouring sites now become correlated. If on-site correlation leads to a charge gap at half-filling, it is certainly a possibility that nearestneighbour correlations for $\delta \approx 0$, for example, might lead to a suppression of the density of states at the chemical potential as well. In fact, it is well documented that all the underdoped cuprates possess a pseudogap[4] in the single-particle spectrum. However, the origin of this phenomenon is unknown. As the pseudogap does not appear to be a true T=0 phase and the pseudogap line joins continuously to the Mott insulator, proposals which require broken symmetry[5, 6, 7, 8] are difficult to reconcile with the Mott state.

Without any assumption as to the nature of the ground state, we show that the the electron spectral function for the 2D Hubbard model contains a dip at the Fermi energy which results in a pseudogap in the single particle density of states (DOS). The pseudogap remains pinned at the Fermi level in the underdoped regime but moves above it at an intermediate doping level, as is seen experimentally [4]. The pseudogap is fundamentally tied to local correlations on neighbouring sites much the way Mott gap arises from on-site physics.

The starting point for our analysis is the Hubbard model with nearest-neighbour hopping matrix element t and on-site Coulomb repulsion U. We base our strong coupling analysis on a two-component composite basis ψ with $\psi_{1\sigma}(i) = \xi_{i\sigma}$ and $\psi_{2\sigma}(i) = \eta_{i\sigma}$ and its associated Green function $S(i,j,t,t') = \langle \langle \psi_{i\sigma}; \psi_{j\sigma}^{\dagger} \rangle \rangle = \theta(t-t')\langle \{\psi_{i\sigma}(t), \psi_{j\sigma}^{\dagger}(t')\} \rangle$, where $\xi_{i\sigma} = c_{i\sigma}(1-n_{i-\sigma})$ and $\eta_{i\sigma} = c_{i\sigma}n_{i-\sigma}$. Here, $c_{i\sigma} = \eta_{i\sigma} + \xi_{i\sigma}$ annihilates an electron on site i and n_i is the number operator for site i. The basis ψ exactly diagonalizes the on-site interaction and hence serves as a natural starting point for a strong-coupling analysis. To overcome the stan-

dard truncation problems inherent in the use of Hubbard operators, we adopt the following procedure. First, project[9] all new operators that arise from the Heisenberg equations of motion of the Hubbard operators onto the Hubbard basis. Second, write the self energy exactly in terms of the remaining operators which are now orthogonal to the Hubbard basis. Third, use local methods in the spirit of dynamical mean-field theory (DMFT)[10] to calculate the resultant electron self energy. To go beyond the single-site treatment indicative of dynamical mean-field theories[10], we adopt the two-site expansion proposed by Mancini and Matsumoto[11] in which the self energy is determined self-consistently from a two-site Hubbard cluster embedded self-consistently in an interacting bath. As all orientations of the two-sites are considered, the electron spectral function will be momentum dependent. Self-consistent cluster methods which are exact as the limit of an infinite cluster appear to be rapidly convergent, providing accurate results for the thermodynamics of the 1D and 2D half-filled bands[9] and in fact constitute the accepted methodology[10] for treating strongly correlated systems. Hence, an implementation of the Hubbard operators coupled with DMFT-type technology places the limitations not on truncation in the equations of motion but on the accuracy of the impurity solver and the size of the finite cluster. As the complete procedure is detailed elsewhere [9, 12], we mention only that in contrast to the work of Matsumoto and Mancini[11], we required that for a fixed filling in the lattice, the chemical potential of the cluster equal that of the lattice.

Using this procedure [9, 12], we report first the doping dependence of the chemical potential. Two distinct possibilities arise: 1) the chemical potential remains pinned and mid-gap states are generated or 2) the chemical potential jumps across the Mott gap. Our results shown in Fig. (1) demonstrate that the chemical potential jumps upon hole or electron doping, indicating an absence of mid-gap states. The Magnitude of the jump is set by the Mott gap which is fully developed at T=0. Even for U = 4t, the inset on the right shows that the chemical potential resides in the LHB for n = 0.95. While at some finite temperature, the chemical potential may appear to evolve smoothly, $\Delta \mu \neq 0$ as the doping increases and hence no mid-gap states are present. Exact results in the 1D Hubbard model[13] as well as Quantum Monte-Carlo simulations[14] in 2D also reveal a chemical potential jump upon doping and hence no mid-gap states. However, unlike 1D and 2D, in $d = \infty$, the chemical potential remains pinned[10] upon doping as midgap states emerge. A chemical potential jump requires a large imaginary part of the self-energy at the chemical potential. From the inset in Fig. (1), we find that $\Im\Sigma$ is initially large in the underdoped regime and acquires the characteristic ω^2 dependence in the overdoped regime indicative of a Fermi liquid. Consequently, the method

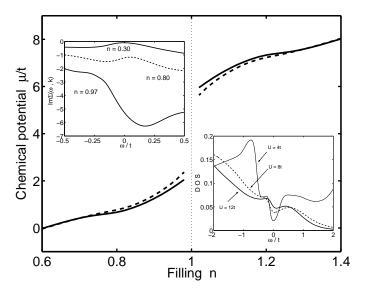


FIG. 1: Doping dependence of the chemical potential in the 2D Hubbard model computed using the local cluster approach for T=0.15t (dashed line) and T=0.07t (solid line). The inset on the left shows the imaginary part of the self energy evaluated at a Fermi momentum (0.3, 2.10) for n=0.97, (0.3, 1.84) for n=0.8 and (0.3, 1.06) for n=0.3, whereas the inset on the right contains the density of states for n=0.95 for U=4t, U=8t, and U=12t.

we use here is capable of recovering Fermi liquid theory in the overdoped regime. Experimentally, whether $\Delta\mu$ vanishes or not appears to be cuprate dependent. For example, in La_{2-x}Sr_xCuO₄[15] (LSCO), the chemical potential remains pinned roughly at 0.4eV above the top of the LHB, while for Nd_{2-x}Ce_xCuO₄ (NDCO)[16] and Bi₂Sr₂Ca_{1-x}R_xCu₂O_{8+y} (BSCO)[17, 18, 19, 20], the chemical potential jumps upon doping and scales roughly as δ^2 as obtained here. Because stripes require $\Delta\mu=0$, they have been invoked[21] to explain the origin of the mid-gap states in LSCO. The pseudogap in the underdoped cuprates has also been attributed[21] to stripes. However, because $\Delta\mu\neq0$ for both NDCO and BSCO, a requirement for stripe formation, if the pseudogap has a universal origin in the cuprates, stripes are not its cause.

To address the origin of the pseudogap, we focus on the doping dependence of the electron spectral function, $-\Im(S_{11}+2S_{12}+S_{22})/\pi)$, shown in Fig. (2). Six features are evident: 1) the chemical potential moves further into the LHB as the filling decreases, 2) no coherent peaks exist near the chemical potential for n=.97, 3) each state in the FBZ has spectral weight both above and below the chemical potential as dictated by Mottness, 4) the Mott gap remains intact but moves to higher energy as the doping increases, 5) at (π,π) , the UHB carries most of the spectral weight regardless of the filling, and 6) a dip exists in the spectral function at the chemical potential for n=0.97 but is absent for n=0.60. In

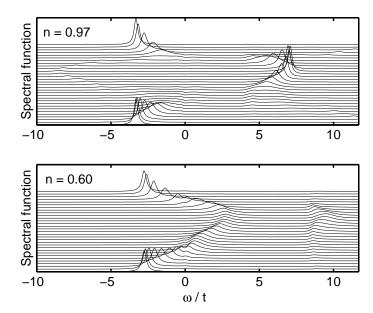


FIG. 2: Electron spectral function for U = 8t, T = 0.07t and n = 0.97 and n = 0.60 along a path in the first Brillouin zone from top to bottom: $(k_x, k_y) = (0, 0) \rightarrow (\pi, \pi) \rightarrow (\pi, 0) \rightarrow (0, 0)$.

the underdoped regime, the characteristic width of each ${\bf k}$ state is of order t and even much larger near $(\pi,0)$. Such broad spectral features in the underdoped regime are seen experimentally[18] and arise in this context because $\Im\Sigma \neq 0$ as shown in Fig. (1). As a consequence, there is no sharp criterion for unit occupancy of each state in the FBZ. Because the spectral weight at each momentum is unity, however, and each state lives both below and above the chemical potential, the charge carried by the piece of the state lying below the chemical potential is less than unity.

Is the dip in the spectral function shown in Fig. (2) for n = 0.97 real? Displayed in Fig. (3) is the DOS for T = 0.25t and T = 0.07t for several fillings. As is evident, no local minimum of DOS exists at the chemical potential at high temperature, T=0.25t. Features which emerge even at high temperature are the reshuffling of spectral weight from above the charge gap to below as the filling is changed and also a movement of the Mott gap to higher energies. Note that even at n = 0.30 the Mott gap is still present, though almost all of the spectral weight now resides in the LHB which closely resembles the non-interacting density of states. This is further evidence that we correctly recover Fermi liquid theory as $n \to 0$. At low temperature, the lower panel of Fig. (3) demonstrates that a pseudogap forms in the DOS for $\delta \approx 0$. The vertical line at 0 indicates that the pseudogap occurs precisely at the chemical potential. Similar qualitative results based on a cluster method have been obtained by Maier, et. al.[22], except their pseudogap is slightly displaced above E_F . In contrast, in the analy-

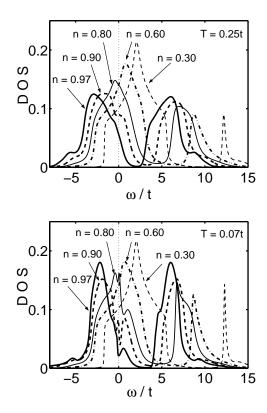


FIG. 3: Density of single particle states for T=0.25t and T=0.07t, U=8t for the fillings shown. No pseudogap exists at high temperature. At low T, a pseudogap emerges and remains pinned at the Fermi level but moves above at an intermediate doping level. In the overdoped regime, the pseudogap vanishes entirely and a non-interacting system is recovered.

sis of Haule, et. al. [23], the DOS has a negative slope through E_F (as dictated by the proximity to the Mott gap) but never acquires a local minimum at E_F indicative of a true pseudogap. Because the pseudogap exists below some characteristic temperature and vanishes at higher doping, the result obtained here is non-trivial and highly reminiscent of experimental trends[4]. What is its origin? The inset in Fig. (1) indicates that for a fixed filling, the pseudogap vanishes as U increases and scales as t^2/U . This suggests that the pseudogap is tied to short-range correlations and hence explains why it is absent in $d = \infty[10]$. While $J_{\text{eff}} \propto O(t^2/U)$ corresponds to the energy scale for antiferromagnetic spin fluctuations, such fluctuations cannot inhibit hole transport. In fact, Maier, et.al.[22] have shown that even if antiferromagnetism is killed, the pseudogap still persists. Further, we have found that J_{eff} is only weakly doping dependent for 0 < x < .25 and in fact vanishes at $x \approx .8$. Hence, the resolution of the pseudogap problem lies elsewhere. The energy scale, t^2/U also describes any transport process in which the intermediate state is doubly occupied. Such processes are captured by our approach as a result of the

coupling of the two-site cluster to the interacting bath. Consider placing a single hole in a Mott insulator. Unlike a site neighbouring the hole, a singly-occupied site two lattice sites away must temporarily doubly occupy one of its neighbours if it is to move to the hole. The energy for this two-step process is t^2/U . Sites further away experience an energy barrier with a higher power of t^2/U . Hence, t^2/U is the largest energy barrier for hole transport once a Mott insulator is doped. Because some sites experience no energy barrier, the single particle density of states exhibits only a suppression, a signal that hole transport involves virtual excitations to the UHB. This pseudogap cannot vanish before each site has on average one hole as its immediate neighbour, roughly x = .25 for a square lattice. Hence, the pseudogap is of the form $t^2/UP(x)$, where P(x) determines the probability that hole transport involves double occupancy and consequently, is a steadily decreasing function of x.

Additionally, it is precisely two-step (or three-site) hopping that makes the single-particle low-energy spectral weight increase faster [3, 25] than 2x. To show that we recover this result, we compute the high and low spectral weight by integrating the DOS from the energy which minimizes the DOS to ∞ ($-\infty$ for electron doping) and from μ to that fixed energy, respectively. The results shown in Fig. (4) (which have been normalized per spin) demonstrate that the initial spectral weight in the UHB which is 1/2 at n=1 all moves to low energies as the filling decreases as is observed experimentally [1, 2]. The same is true for electron doping (n > 1). Further, the curvature of the low energy spectral weight (LESW) is positive indicating that the LESW grows faster[3, 25] than 2x. The growth in excess of 2x arises entirely from virtual excitations between the LHB and UHB and points to an iseparability of the low and high energy scales. Such behavior is absent from a band insulator (see W_{NI} in Fig. (4). That Mottness leads to such a drastic deviation from the non-interacting result is a direct consequence of each state having spectral weight both above and below the chemical potential (see Fig. (2)). Our finding that the three-site terms lead to an inseparability of low and high-energy scales resonates with the recent work of Kirkpatrick and Belitz[26] who have shown that three-body terms are ubiqutious in strongly correlated electron systems and lead to breakdown of a true lowenergy description.

Without global symmetry breaking, spin-charge separation, or pairing, we have shown that lightly doped Mott insulators posess a pseudogap which arises entirely from nearest-neighbour correlations. The pseudogap is ubituitous because any singly-occupied site not immediately neighbouring a hole experiences an energy gap for transport equal to t^2/U . This generic phenomenon offers a simple resolution of the pseudogap problem in the cuprates.

This work was funded by the Petroleum Research Fund

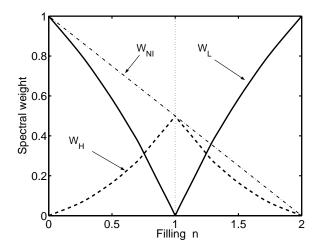


FIG. 4: High (W_H) and low (W_L) spectral weight as a function of filling. W_{NI} is the spectral weight in the non-interacting system.

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